BROADBAND LIGHT SCATTERING AND ANOMALOUS RELAXATIONS OF SUPERCOOLED PROPYLENE GLYCOL

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Most supercooled liquids transform into glassy states, while the mechanism of this liquid-glass transition is still unknown. Thus the dynamical properties of glass-forming liquids are today a subject of increasing interest in connection with the poorly understood liquid-glass transition phenomena.

In the present study, the dynamical properties were studied in propylene glycol. It undergoes a liquid-glass transition at about 172K. It belongs to an intermediate liquid according the classification of strong and fragile liquids. The low frequency relaxation has been already studied by dielectric and photoacoustic measurements.[1] Therefore the higher frequency region above 1GHz was studied by the broadband light scattering experiments.

The propylene glycol sample with 99.5% purity was purchased from Wako Pure Chemical Industries, Ltd. It was distilled and desiccated prior to use. The broadband spectra were obtained from 1GHz to 10THz, where the region below 500GHz was measured utilizing the Sandercook=type Fabry-Perot interferometer.[2] The region above 100GHz was measured using the triple-grating spectrometer of additive dispersion (Jobin Yvon T64000). The output signals were detected by the photon counting system in both cases.

The temperature dependence of the low-frequency polarized Raman scattering spectra was already reported.[3] With the decrease of temperature intense Rayleigh wing reduces markedly and the boson peak appears. This is one of the typical nature of liquid-glass transitions, which has been observed in various glass-forming materials.

The dynamical susceptibility was determined from the depolarized scattering spectra in a large frequency range between 1GHz and 10THz by the combination of Brillouin and Raman scattering measurements. The obtained spectra were analyzed by the mode coupling theory at first, however it is found that the agreement is not sufficient as same as the case of glycerol.[6] Next the phenomenological approach was attempted by the three contributions, namely α-relaxation, fast relaxation and boson peak. Figure 1 shows the combined broadband spectra and calculated results. The spectrum at 363K reveals the two peaks as shown in Fig.1(a). With the decrease of temperature the lower α peak shifts to more lower frequency, while the boson peak does not change remarkably as shown in Fig.1 (b). The lower peak is attributed to α relaxation. According to the previous experimental study of the dielectric and the thermal dispersion, α relaxation is well reproduced by the Davidson-Cole formula, and its relaxation frequency obeys the Vogel-Tammann-Fulcher law.[1] The higher peak corresponds to the boson peak. The previous study by the Raman scattering shows that the lineshape of the boson peak is well fitted by the disorder-induced model with the exponential correlation function.[4] However ,in fact, the another contribution is necessary to reproduce the middle region between two peaks. Thus the another relaxation mode of the Debye type was introduced. Consequently the observed spectra were well reproduced by these three contributions.

REFERENCES

1) S.Kojima: Ferroelectrics : 137 (1993) 315.
2) A.Yoshihara: Jpn. J. Appl. Phys. 33 (1994) 3100.
3) S.Kojima: Jpn. J. Appl. Phys. 32 (1993) 2223.
4) S.Kojima:Phys. Rev. B47 (1993) 2924.

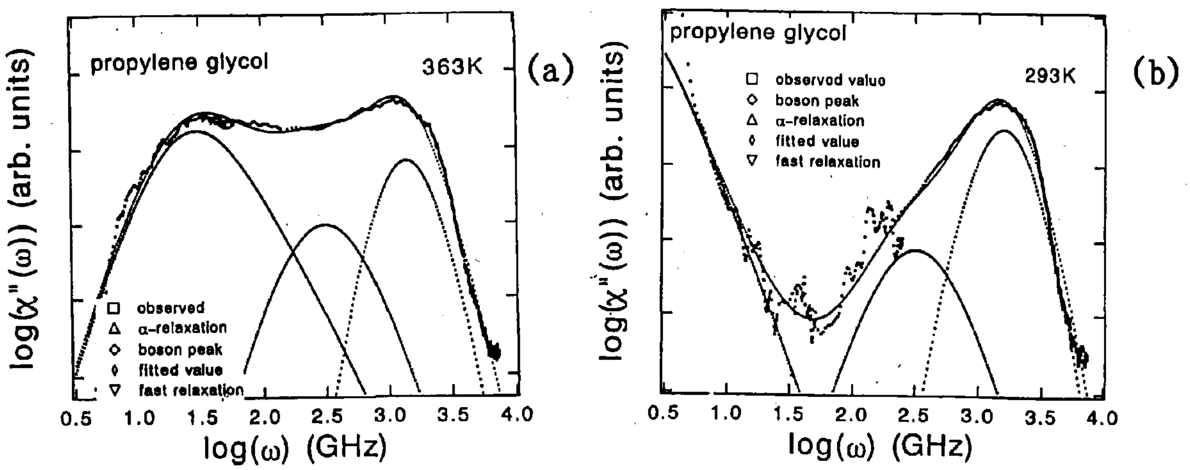

Fig.1 Broadband dynamical susceptibility obtained by the connection of Brillouin and Raman scattering spectra. (a) 363K, (b) 293K.